

US EPA ARCHIVE DOCUMENT

# CMAQ Tagged Species Source Apportionment (TSSA)

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# Motivation

- Need to understand which emissions sources contribute to haze and other pollutants.
- Use this information to assist in developing control strategies.

# **Modeling Approaches for Source Apportionment**

- Chemical Mass Balance analysis
- PCA and Positive Matrix Factorization
- Back-trajectory models, eg, HYSPLIT
- Lagrangian or Particle models, eg, CALPUFF
- Grid Model Sensitivity Studies
- Grid Model Tracer Approaches.

# Sensitivity Methods (1)

Brute force approach:

1. Run a model Base Case simulation.
2. Add-in or Zero-out a particular source and run a the model again.
3. The difference in the base case and the sensitivity case predicts the effect of changing that emissions source on air pollutant concentrations at all receptor sites.

## Sensitivity Methods (2)

- Advantages of Sensitivity Methods:
  - directly related to development of control measures.
  - Conceptually simple to apply.
  - Accurate for species with linear chemistry and for small (2 to 20%) changes in emissions.

## Sensitivity Methods (3)

- Problems with Sensitivity Methods:
  - Brute force approaches (removing one species in each model run) are computationally expensive.
  - Sensitivity results depend on the base case scenario (clean versus polluted base case).
  - Sensitivity results can be non-linear and non-additive, especially for high NO<sub>x</sub> conditions.
  - CMAQ exhibits numerical noise in PM sensitivity runs.

# Grid Model Tracer Methods

- New tracers or tags and new algorithms are added to air quality models to track the chemical conversions, transport and fate of selected emissions sources:
  - CMAQ Tagged Species Source Apportionment (TSSA).
  - CAMx PM Source Apportionment Technology (PSAT).
  - REMSAD also has a source apportionment algorithm.



# CMAQ TSSA Objectives

- Use “Tagged Species” or tracers to track the chemical transformations, transport and loss of each PM species and each PM precursor .
- Add the tracers for key species and for defined source regions & source categories.
- Provides 3-D fields showing source attribution of of PM species for each grid cell in model domain.
- Provides full attribution of PM at any any receptor site to individual emissions source groups.

# CMAQ TSSA Approach

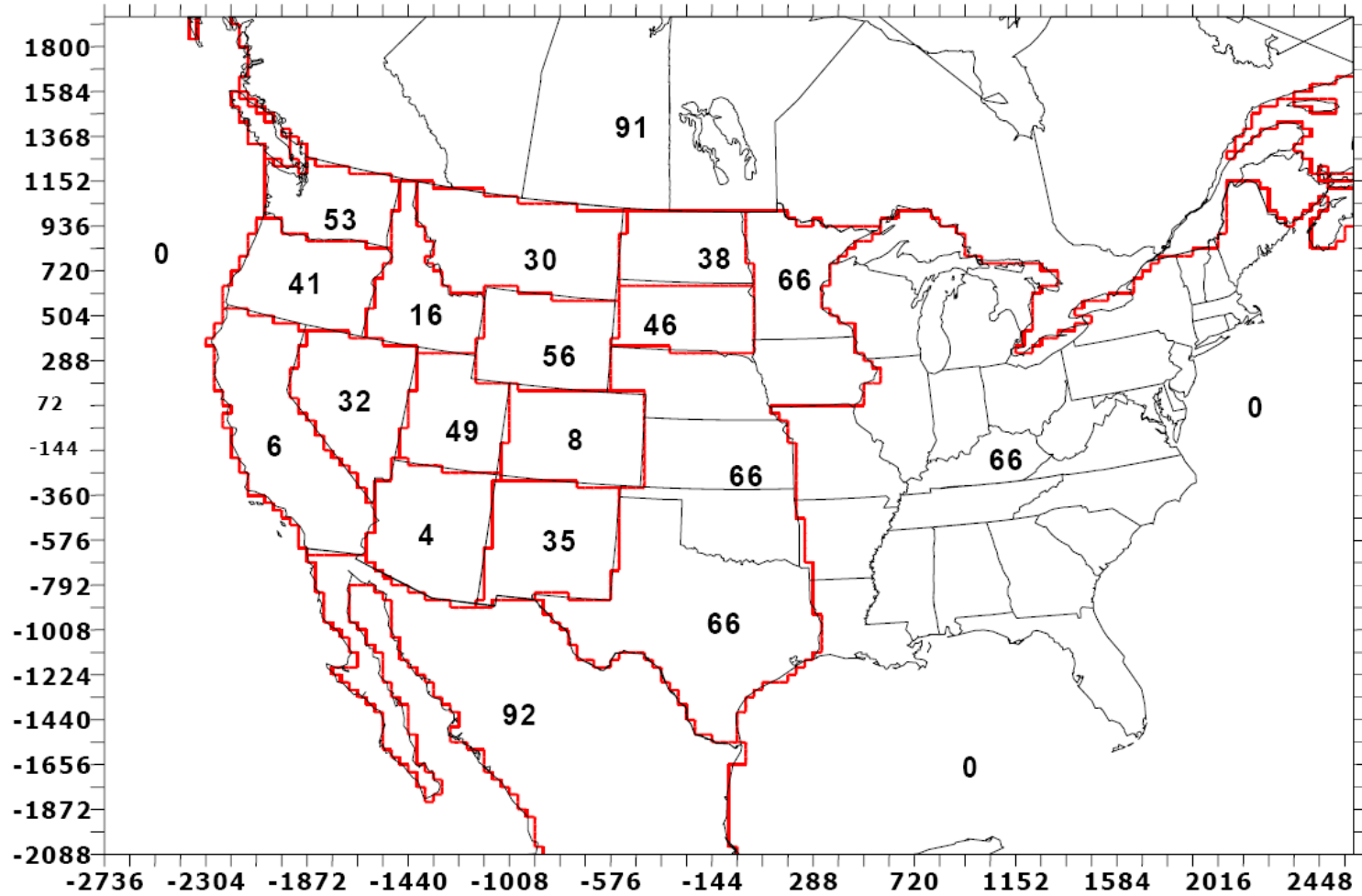
- Method: Add one new set of tagged species for each emissions source category or source location being tracked.
  - Straight forward for non-reactive species: add 1 tracer for each source.
  - Example: in each grid cell the sum of all tracers for EC equals the total (bulk) EC concentration in each cell.
  - Each tracer is defined for all grid cells and is emitted, transported, and removed proportional to its weight of the bulk species in each grid cell.

## CMAQ TSSA Approach (2)

- More complicated for chemical reactive species and secondary particulates:
  - Must also track the chemical reactions that convert a tracer between different gas species and from gas to PM.
  - Model include approximately 6 forms of N species, must carry 6 additional tracers for each NO<sub>x</sub> source category to track the contributions to aerosol nitrate.
  - SOA formation is still more complex, not included in current CMAQ algorithm.
  - SO<sub>4</sub> and other PM species are easier and less computational expensive to treat.

# Source areas defined by numerical codes for domain of interest.

TSSA MAP



(1) Source Regions

# Emissions Source Tags

Types	Source Category	Notes
ICON	ICON	Initial Concentration
BCON	BCON	Boundary Concentration
Emissions	MV_*	Mobile sources of any state
	BG_*	Biogenic sources of any state
	RD_*	Road dust of any state
	NR_*	Non- Road dust of any state
	PN_*	Point sources of any state
	AR_*	Area sources of any state
	WF_*	Wild Fire of any state
	AG_*	Ag burning of any state
	RX_*	Prescribed burning of any state
	ET_*	Total Emission of any state
	*_WRAP	Any type of emission of WRAP states
	*_VISTAS	Any type of emission of WRAP states
Others	OTHERS	Any sources other than all of the above

# CCTM Governing Equation

$$\frac{\partial(\bar{\varphi}_i J_\xi)}{\partial t} + m^2 \nabla_\xi \cdot \left( \frac{\bar{\varphi}_i \bar{\mathbf{V}}_\xi J_\xi}{m^2} \right) + \frac{\partial(\bar{\varphi}_i \bar{v}^3 J_\xi)}{\partial \hat{x}^3}$$

(a) (b) (c)

$$-m^2 \frac{\partial}{\partial \hat{x}^1} \left[ \frac{\bar{\rho} J_\xi}{m^2} (\hat{K}^{11} \frac{\partial \bar{q}_i}{\partial \hat{x}^1}) \right] - m^2 \frac{\partial}{\partial \hat{x}^2} \left[ \frac{\bar{\rho} J_\xi}{m^2} (\hat{K}^{22} \frac{\partial \bar{q}_i}{\partial \hat{x}^2}) \right] - \frac{\partial}{\partial \hat{x}^3} \left[ \bar{\rho} J_\xi (\hat{K}^{33} \frac{\partial \bar{q}_i}{\partial \hat{x}^3}) \right]$$

(d) (e)

$$-m^2 \frac{\partial}{\partial \hat{x}^1} \left[ \frac{\bar{\rho} J_\xi}{m^2} (\hat{K}^{13} \frac{\partial \bar{q}_i}{\partial \hat{x}^3}) \right] - m^2 \frac{\partial}{\partial \hat{x}^2} \left[ \frac{\bar{\rho} J_\xi}{m^2} (\hat{K}^{23} \frac{\partial \bar{q}_i}{\partial \hat{x}^3}) \right]$$

(f)

$$-\frac{\partial}{\partial \hat{x}^3} \left[ \bar{\rho} J_\xi (\hat{K}^{31} \frac{\partial \bar{q}_i}{\partial \hat{x}^1} + \hat{K}^{32} \frac{\partial \bar{q}_i}{\partial \hat{x}^2}) \right]$$

(g)

$$= J_\xi R_{\varphi_i}(\bar{\varphi}_1, \dots, \bar{\varphi}_N) + J_\xi Q_{\varphi_i} + \frac{\partial(\bar{\varphi}_i J_\xi)}{\partial t} \Big|_{\text{cld}} + \frac{\partial(\bar{\varphi}_i J_\xi)}{\partial t} \Big|_{\text{aero}} + \frac{\partial(\bar{\varphi}_i J_\xi)}{\partial t} \Big|_{\text{ping}} \quad (6-14)$$

(h) (i) (j) (k) (l)

- a. Time rate of change in species concentration;
- b. Horizontal advection;
- c. Vertical advection;
- d. Horizontal eddy diffusion (diagonal term);
- e. Vertical eddy diffusion (diagonal term);
- f. Off-diagonal horizontal eddy diffusion;
- g. Off-diagonal vertical eddy diffusion;
- h. Production & loss from chemical reactions;
- i. Emissions;
- j. Cloud mixing and aqueous-phase chemical production & loss;
- k. Aerosol process; and
- l. Plume-in-grid process

# CCTM Governing Equations

- Chemistry:  $N$  coupled PDEs

$$\partial C_j / \partial t + \mathbf{v} \cdot \nabla C_j = \mathbf{D} \nabla^2 C_j + P - L C_j + E_j - D_j, \quad j=1, N$$

- Operator Splitting: Solve each term independently for a short time step:

$$\partial C_j / \partial t = - \mathbf{v} \cdot \nabla C_j \quad \text{Advection}$$

$$\partial C_j / \partial t = \gamma \nabla^2 C_j + E_j - D_j \quad \text{Diffusion, Emis, Depo}$$

$$dC_j / dt = P - L C_j \quad j=1, N \quad \text{Chemistry}$$

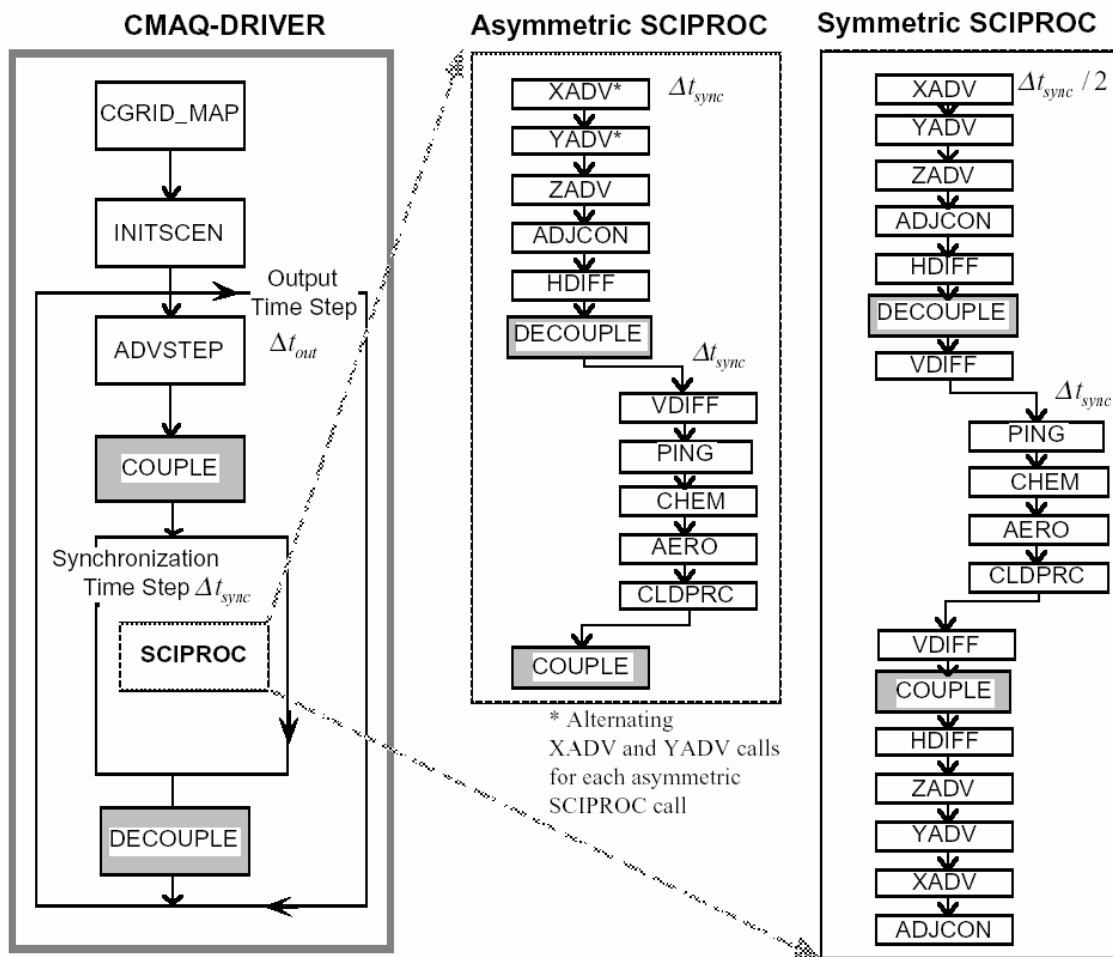
# CMAQ Chemical Transport Model (CCTM) Science Modules

- Currently, nine science modules are included:
  - **DRIVER** controls model data flows and synchronizes fractional time steps;
  - **HADV** computes the effects of horizontal advection;
  - **VADV** computes the effects of vertical advection;
  - **ADJCON** adjusts mixing ratio conservation property of advection processes;
  - **HDIFF** computes the effects of horizontal diffusion;
  - **VDIFF** computes the effects of vertical diffusion and deposition;
  - **CHEM** computes the effects of gas-phase chemical reactions;
  - **CLOUD** computes the effects of aqueous-phase reactions and cloud mixing;
  - **AERO** computes aerosol dynamics and size distributions; and
  - **PING** computes the effects of plume chemistry.

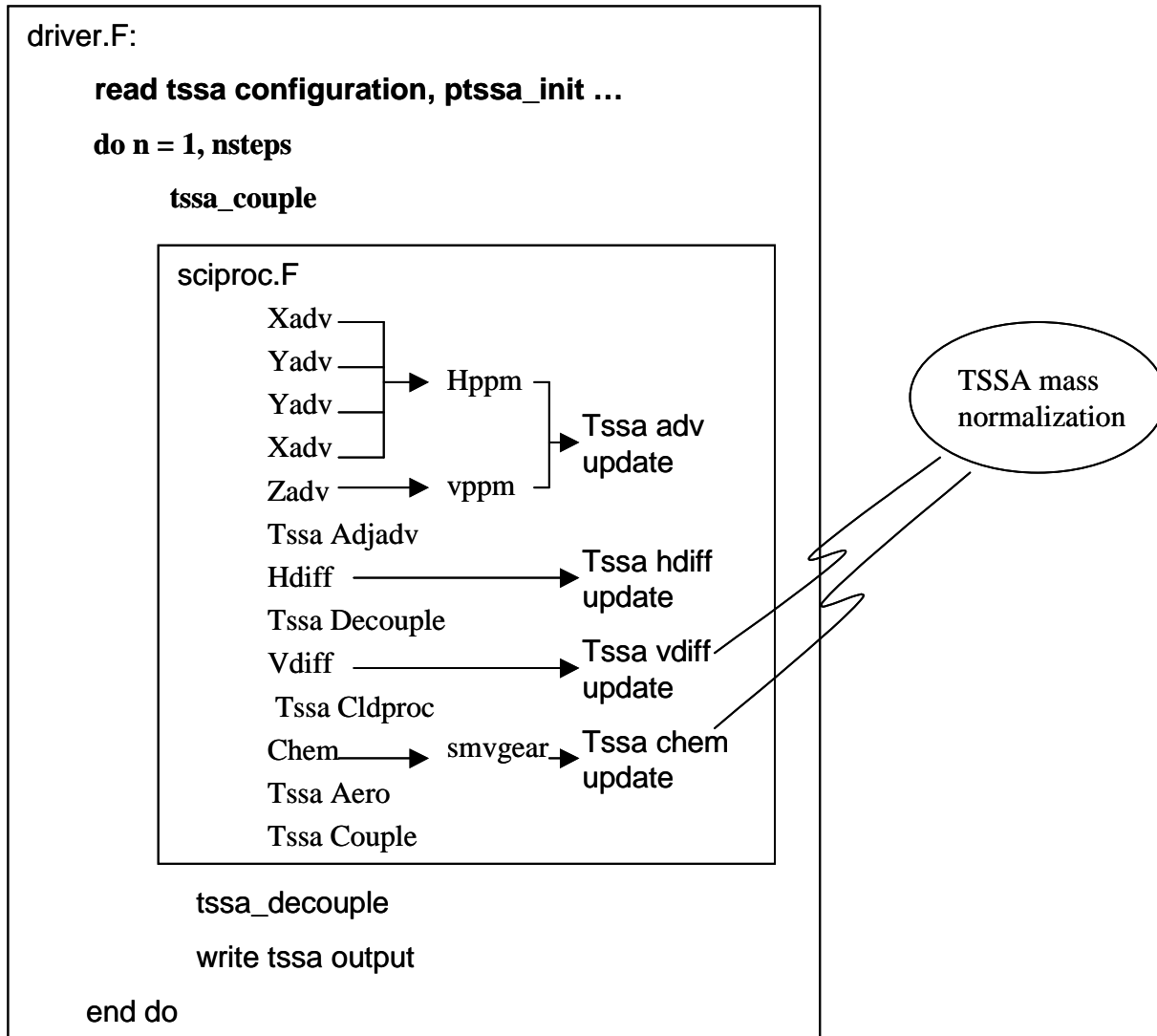


# CCTM

## Driver Module and Science Processor



# TSSA Flow Diagram



# Initial and Boundary Conditions

- Tracer initial condition:
  - the tagged species are initialized with concentrations from the model initial condition.
  - Requires model spin-up period of 2 to 15 days depending on domain size.
- Boundary conditions:
  - The TSSA algorithm updates the tagged species at the boundary using concentrations from the boundary conditions input file, when there is influx to the model domain.
  - Outflux at the boundaries is treated as simple removal process (no recirculation of mass back into the domain).

# Transport & Loss Terms

- Advection
  - Calculate mass flux between grid cells to update tracers.
- Vertical Diffusion
  - Apply the CMAQ diffusion algorithms to tracers.
  - evaluated algorithms to estimate actual 2-way mass transfer between layers, but problems with this approach.
- Update for mass flux in CLOUD & aqueous chemistry algorithm.
- Update tagged species for emissions and deposition terms.
- Check for mass conservation at each step and adjust mass if needed. Halt if large errors.

# Horizontal and Vertical Advection

- The TSSA algorithm uses CCTM's transport solver to advect each tracer species. The fluxes calculated at the upwind and downwind grid cell boundaries are used to update each tracer species.
- TSSA is currently implemented into the Piecewise Parabolic Method (PPM) and Yarmatino schemes.
- Mass Adjustment step renormalizes sum of tracers if it diverges from bulk concentration.

# Vertical Diffusion

- A semi-implicit scheme is used in CMAQ to solve vertical diffusion:
  - The net change in species concentration at each layer is obtained by solving the tri-diagonal matrix with species concentration from the current and the next time steps.
- The tracer tagged-species is solved using the CMAQ diffusion algorithms to calculate the net mass exchange of each tracer species.

# Deposition

- Coupled with vertical diffusion in CMAQ.
- The TSSA tagged-species are deposited the same way as the CMAQ species,
  - with the loss of each tagged-species being proportional to the deposition rate of the bulk species, and
  - the sum of deposition for all tagged-species, including “other”, summing to equal the deposition of the bulk species

# Emissions

- Emissions are treated as a simple source that increments the bulk species:
  - If a given emissions source is selected for the TSSA algorithm, the tagged-species is also incremented.
  - Emissions sources that are not selected for the TSSA are added to a tagged-species identified as “other”.
  - This approach allows for all emissions to be included in the TSSA algorithm
- The tagged-species NOX is incremented by emissions of all reactive N species, i.e., NO, NO<sub>2</sub> and HONO emissions.



# Chemistry

- The integrated rates of chemical reactions are used to calculate the mass transfer from one bulk species to another at each chemistry time step (requires use of IRR option in CMAQ)
- In the simplest case for a species with chemical loss but no chemical production, simply reduce the tagged-species proportionally to the reduction in the bulk species.
- In the more complex case in which there is both chemical production and loss of species  $C_j$ , the rate of removal of the bulk and tagged components is calculated by solving a simple ordinary differential equation.

$$\frac{dC_{j,bulk}}{dt} = P_{j,Total} - LC_{j,bulk}$$

# Tagged Species for Nitrates

- NOX = reactive N family.  
= { NO, NO<sub>2</sub>, NO<sub>3</sub>, 2\*N<sub>2</sub>O<sub>5</sub>, HONO, PNA }
- HNO<sub>3</sub>
- PAN
- RNO<sub>3</sub>
- ANO<sub>3</sub>J
- ANO<sub>3</sub>I

# Chemical Transformations

- Emissions are as  $\text{NO}_x = \text{NO} + \text{NO}_2$
- Use integrated reaction rates at each time step to update the tagged species:
  - $\text{NO}_x \leftrightarrow \text{PAN}$
  - $\text{NO}_x \leftrightarrow \text{RNO}_3$
  - $\text{NO}_x \rightarrow \text{HNO}_3$
  - $\text{HNO}_3 \leftrightarrow \text{ANO}_3$

# Aerosols in TSSA

- At each step the CMAQ algorithms where aerosol species are updated, the tagged-species are also updated with the change being proportional to the change in the bulk species concentration at each time-step in the aerosol solver.
- For mass transfer between gas and aerosol phase we assume that all tagged species gas and aerosol species are in thermodynamic equilibrium.

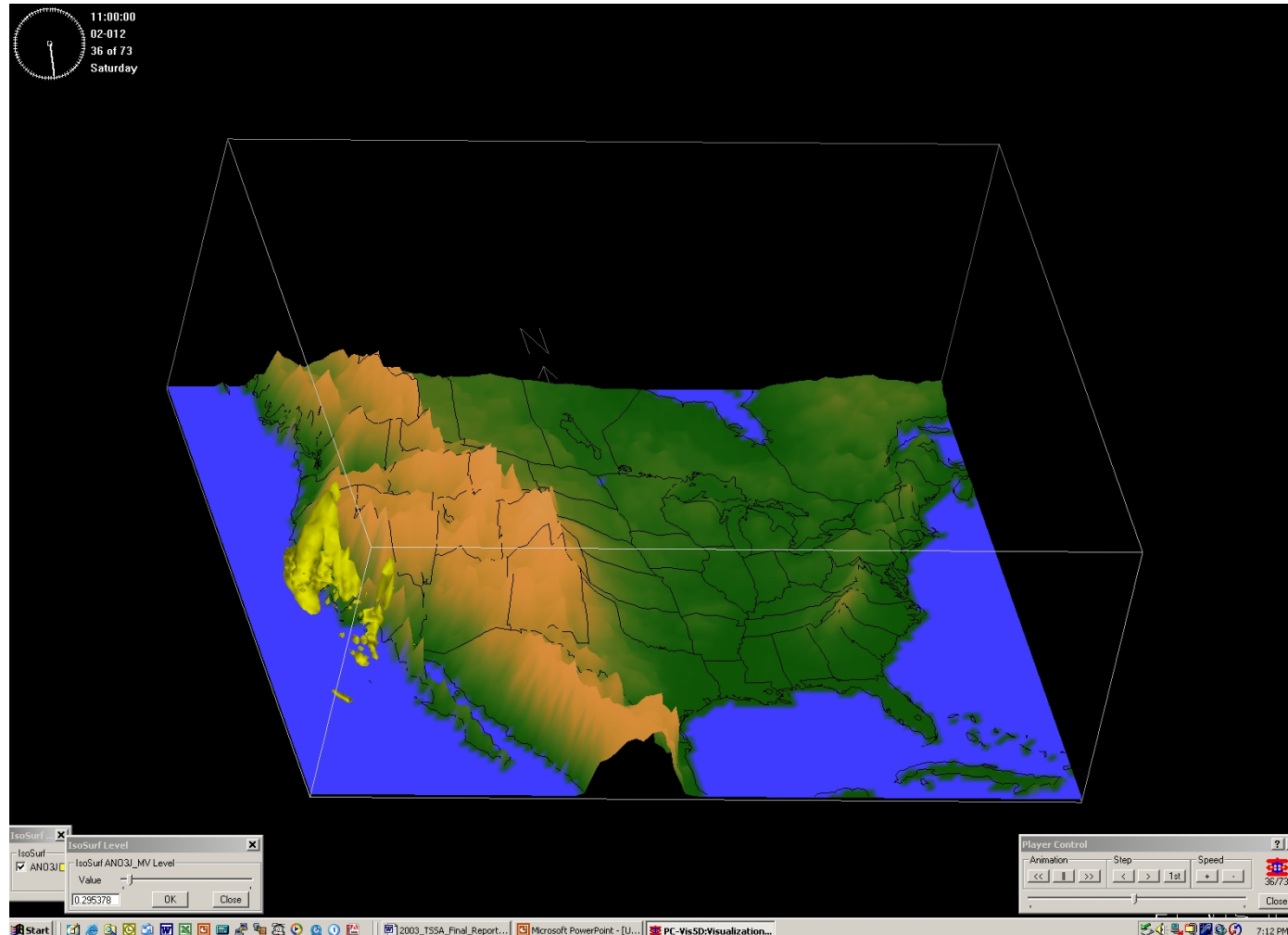
# Implementation in CMAQ

- First implemented in CMAQ v4.2.2. (12/03) and CMAQ v4.4 beta (4/04). Problems with mass conservation in CMAQ v4.x.
- Currently implemented in CMAQ v4.5 with GEAR & QSSA chemistry solvers.
- Parallelism: supports multi-processor usage (MPI)

# Output Formats

- 3-D concentration field for each tagged species
  - Each model grid cell is included as a receptor.
  - Results can be viewed as 3-d animation of the tagged species, or layer 1 animations in PAVE
  - Bar plots for receptor showing attribution is most useful for the receptor sites.
- Output files for annual simulation are too large with over 12 GB per day.
  - Currently outputting only layer 1.

# Aerosol Nitrate from CA Mobile Emissions



# Computational Cost/Constraints

- Slower run time because we use GEAR or QSSA chemistry (about 4x cost of EBI solver).
- RAM memory is primary constraint:
  - For many tracers we split them between 2 model simulations.
- Run Time: about 4.5 hours per day using 8 Opteron CPUs with 2 GB RAM.
- With 32 CPUs, running CMAQ in 4 seasons, takes about 2 weeks for an annual simulation.



# Testing and Validation

- Compared CMAQ/TSSA to CMAQ.
- Compared tracers to model zero-out sensitivity simulations.
- Compared with CAMx/PSAT
- Not possible to compare with HYSPLIT back trajectory modeling - temporal scales not consistent.

# Post-processing Analysis

- PAVE spatial plots showing region of influence of a given source (at a point in time or averaged over some time period).
- Vis5d animations showing plume from a source.
- Bar plots showing all sources that contribute to PM at a given receptor site.

# BC Tracer test: CO from boundaries

## CMAQ/TSSA

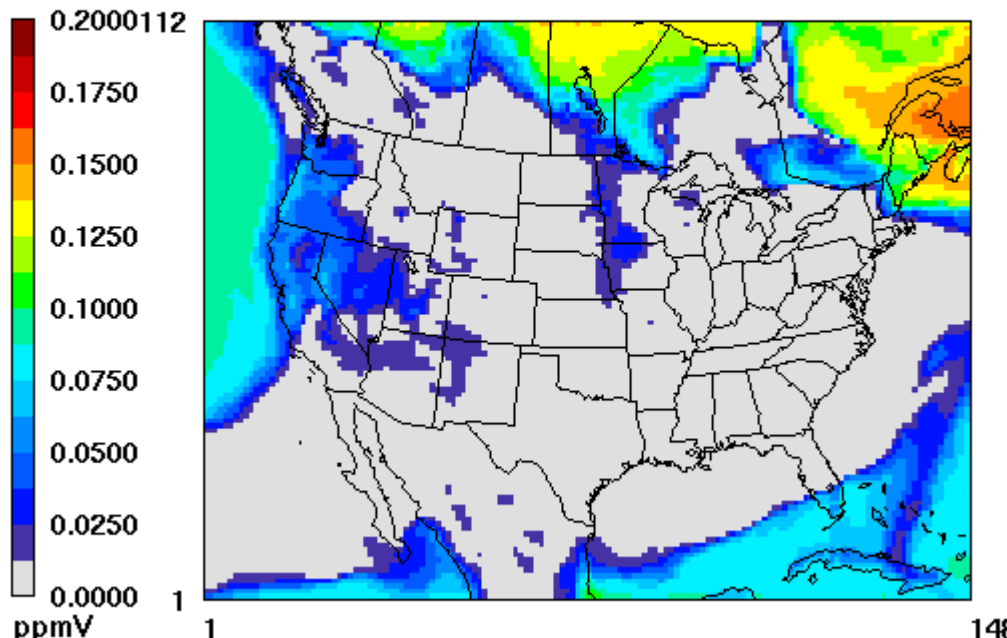
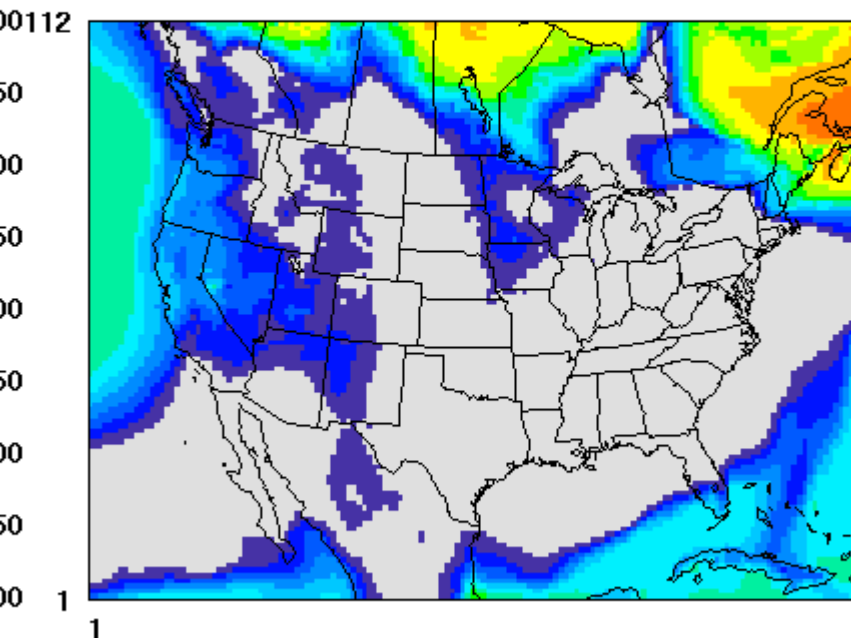
## Zero-out Sensitivity

CO\_BCON

CO

CMAQ\_TSSA CO\_BCON  
WRAP Base18a CMAQ v4.5 ppm

CONC Diff: BCON zero-out run  
WRAP Base18a CMAQv4.5 ppm



June 17,2002 0:00:00  
Min= 0.0000 at (98,29), Max= 0.1592 at (148,90)

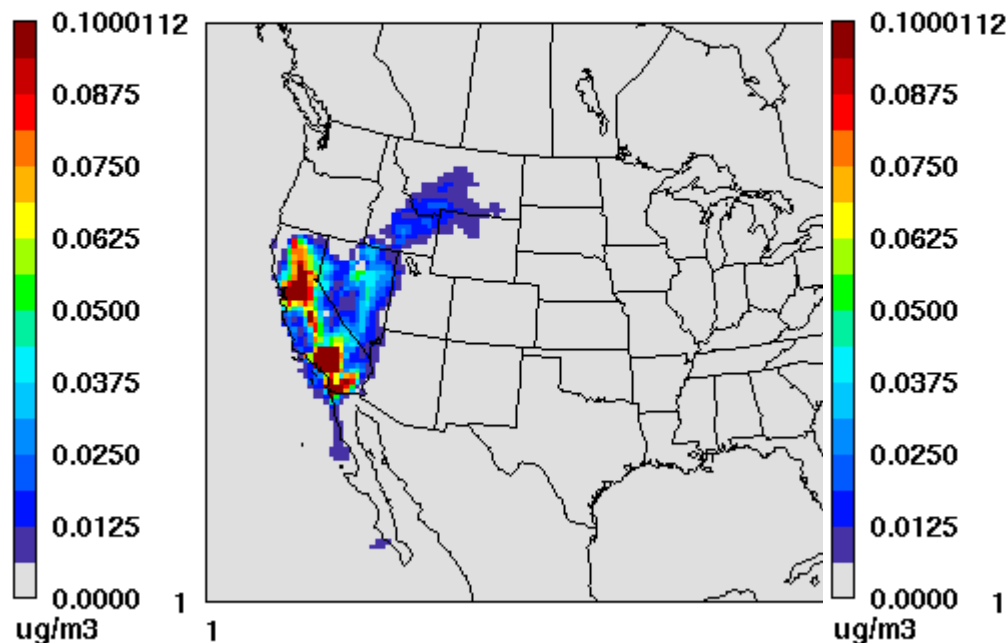
June 17,2002 0:00:00  
Min=-0.0047 at (138,72), Max= 0.1591 at (148,90)

# Non-reactive Tracer test: EC mobile sources

## CMAQ/TSSA

AEC\_MV\_CA

CMAQ\_TSSA AEC\_MV\_CA  
WRAP Base18a CMAQ v4.5 ppm

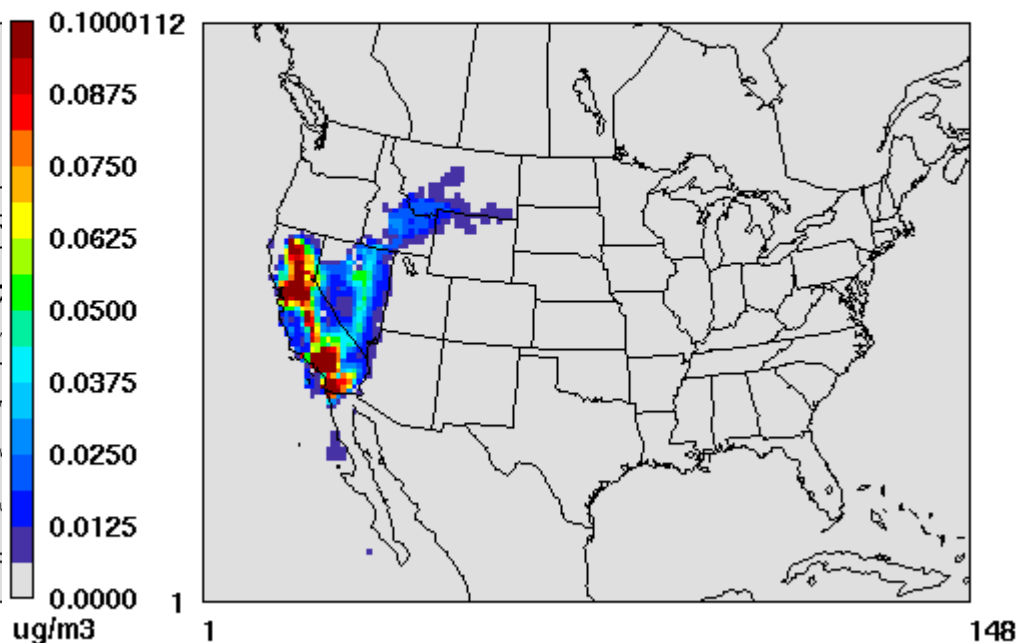


June 22,2002 0:00:00  
Min= 0.0000 at (148,12), Max= 0.4840 at (

## Zero-out Sensitivity

AEC

CONC Diff: CA EC zero-out run  
WRAP Base18a CMAQv4.5 ppm



June 22,2002 0:00:00  
Min=-0.0118 at (22,45), Max= 0.5009 at (23,46)

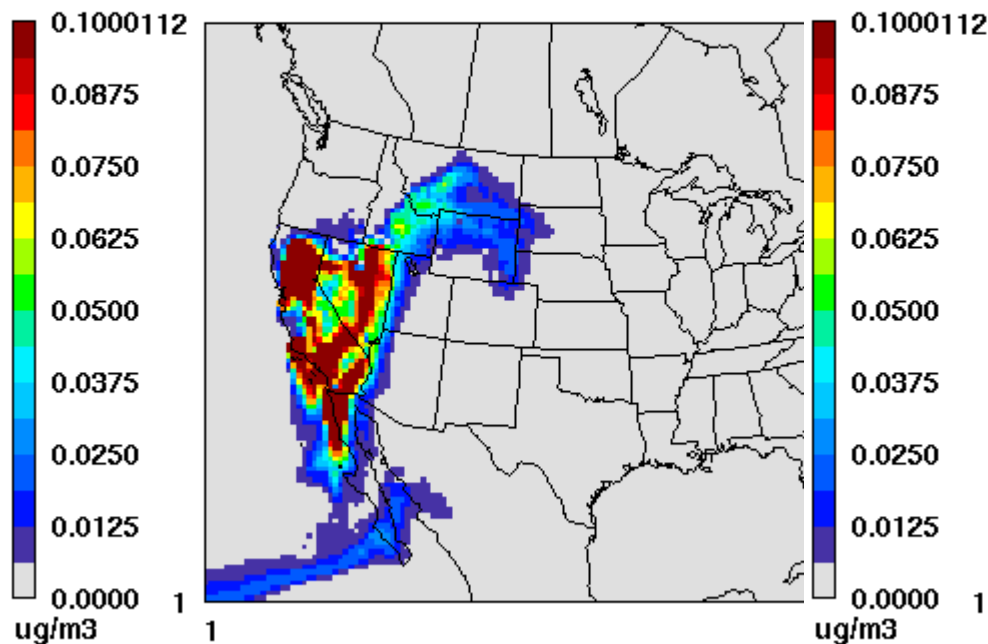
# Reactive Tracer test: Sulfate

## SOx point sources from CA

### CMAQ/TSSA      Zero-out Sensitivity

ASO4\_PT\_CA

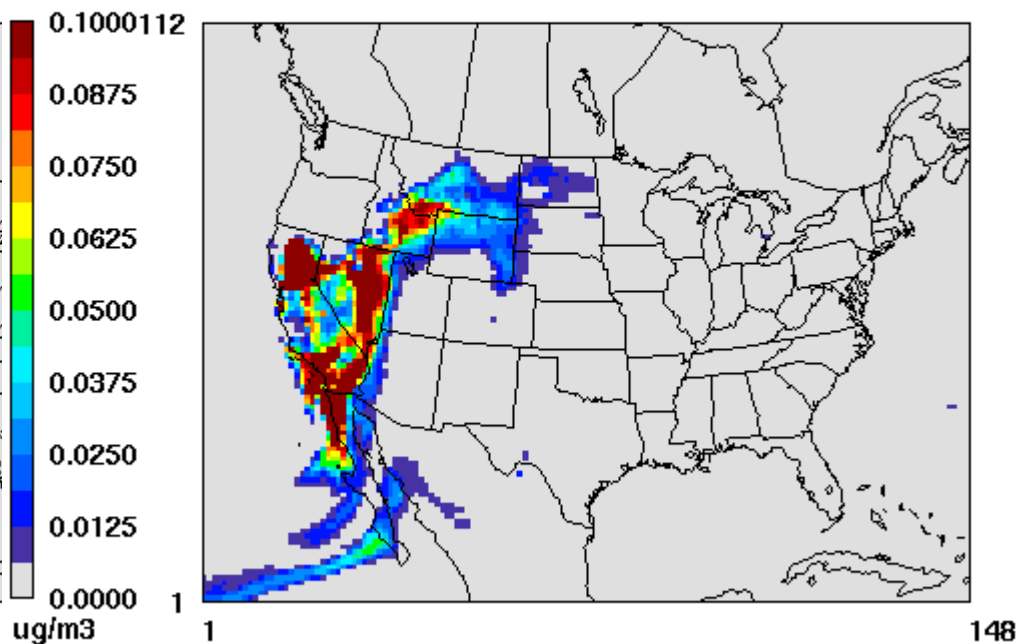
CMAQ\_TSSA ASO4\_PT\_CA  
WRAP Base18a CMAQ v4.5 ppm



June 22, 2002 0:00:00  
Min= 0.0000 at (148,27), Max= 1.5824 at

ASO4

CONC Diff: CA SOX zero-out run  
WRAP Base18a CMAQv4.5 ppm



June 22, 2002 0:00:00  
Min=-0.0518 at (15,65), Max= 1.8821 at (25,39)

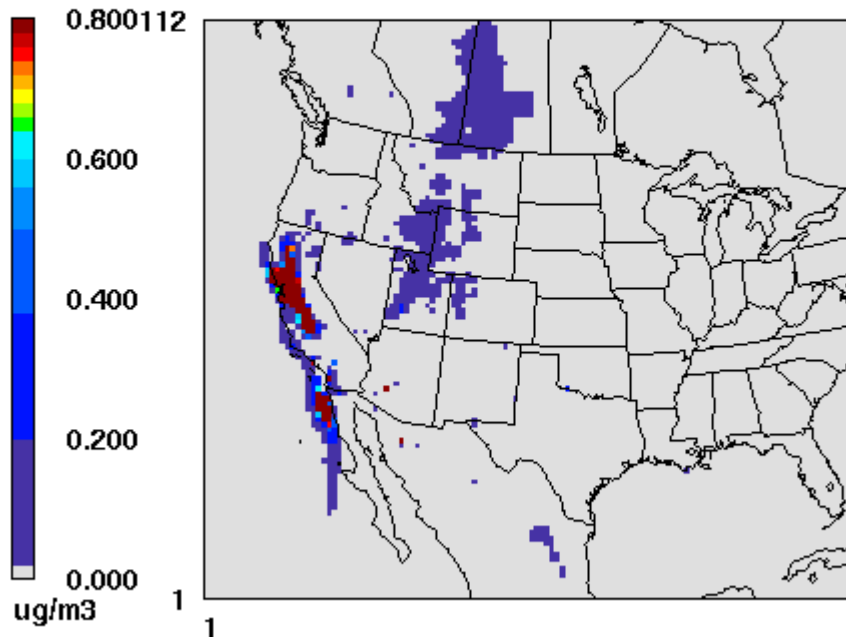
# Non-reactive Tracer test: Nitrate

## NO<sub>x</sub> mobile sources from CA

### CMAQ/TSSA      Zero-out Sensitivity

ANO3

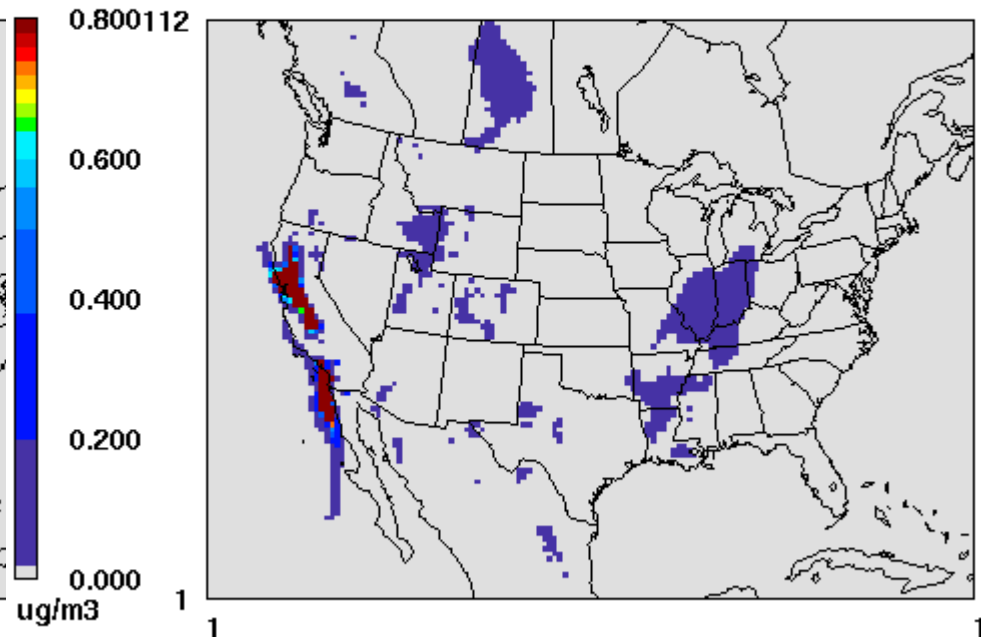
ACONC Diff: CA Mobile NO<sub>x</sub> zero-out ru  
WRAP Base18a CMAQv4.5 ppm



January 7, 2002 0:00:00  
Min= -0.997 at (24,45), Max= 3.822 at (19,

ANO3\_MV\_CA

CMAQ\_TSSA ANO3\_MV\_CA  
WRAP Plan02b CMAQ v4.5 ppm



January 7, 2002 0:00:00  
Min= 0.000 at (1,90), Max= 3.422 at (24,43)

# Comparison of TSSA and PSAT

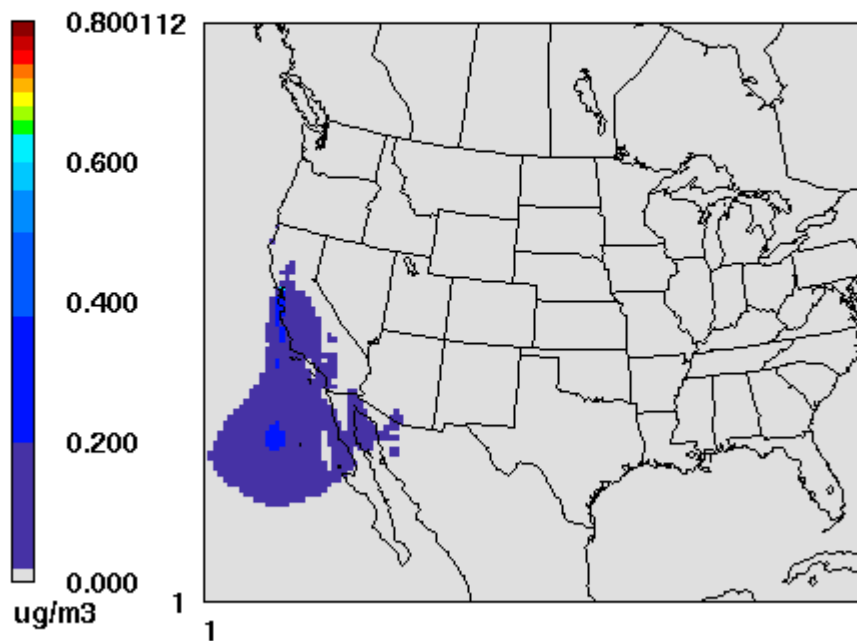
- CMAQ/TSSA and CAMx/PSAT were applied for the same model scenarios using identical MM5 and emissions input data.
- Both models use similar approach for source apportionment, expect to see similar results.
- Difference in models' advection & dispersion cause differences in the models' predicted concentrations, but we expect spatial patterns to be similar.

# Comparison of CMAQ and CAMx: Sulfate SO<sub>x</sub> point sources from CA

## CMAQ/TSSA                      CAMx/PSAT

ASO4\_PT\_CA

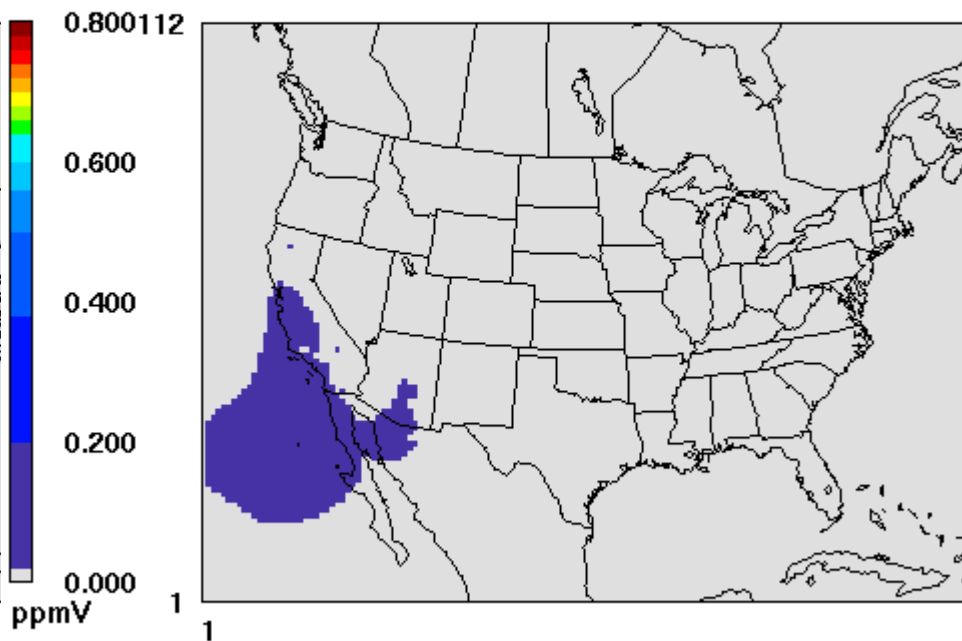
CMAQ\_TSSA ASO4\_PT\_CA  
WRAP Plan02c CMAQ v4.5 ppm



January 31,2002 0:00:00  
Min= 0.000 at (1,110), Max= 0.611 at (16,1)

PSO4 Point Source CA

CAMx Planning 02c



January 31,2002 0:00:00  
Min= 0.000 at (1,1), Max= 0.161 at (16,61)



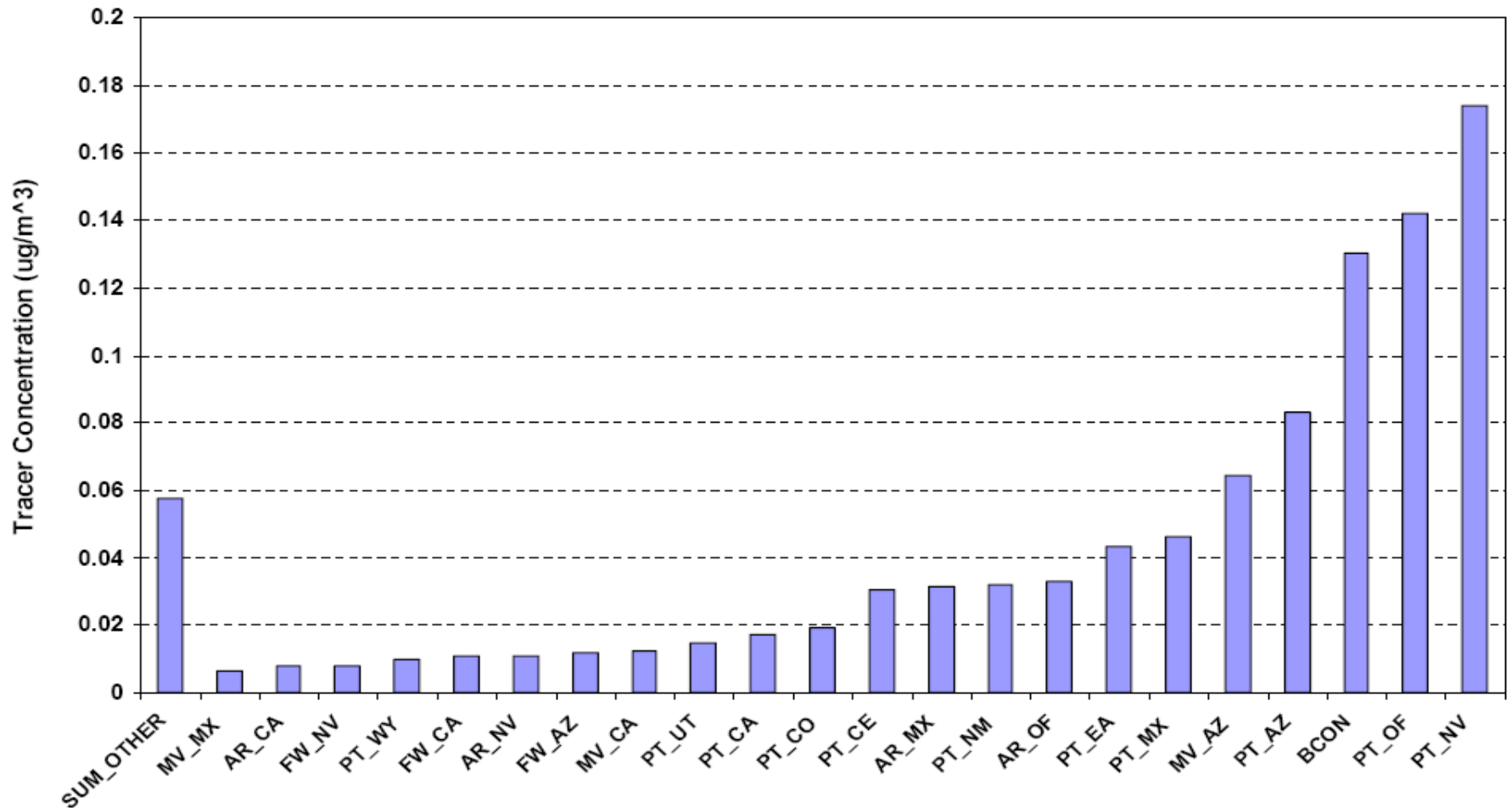
# TSSA Sulfate Contributions at a Receptor Site

## Hance Camp at Grand Canyon NP, AZ (GRCA2)

WRAP Plan02c TSSA Tracers of ASO4: July

Sum of all TRACER Concentration = 0.9946

ACONC = 0.9946

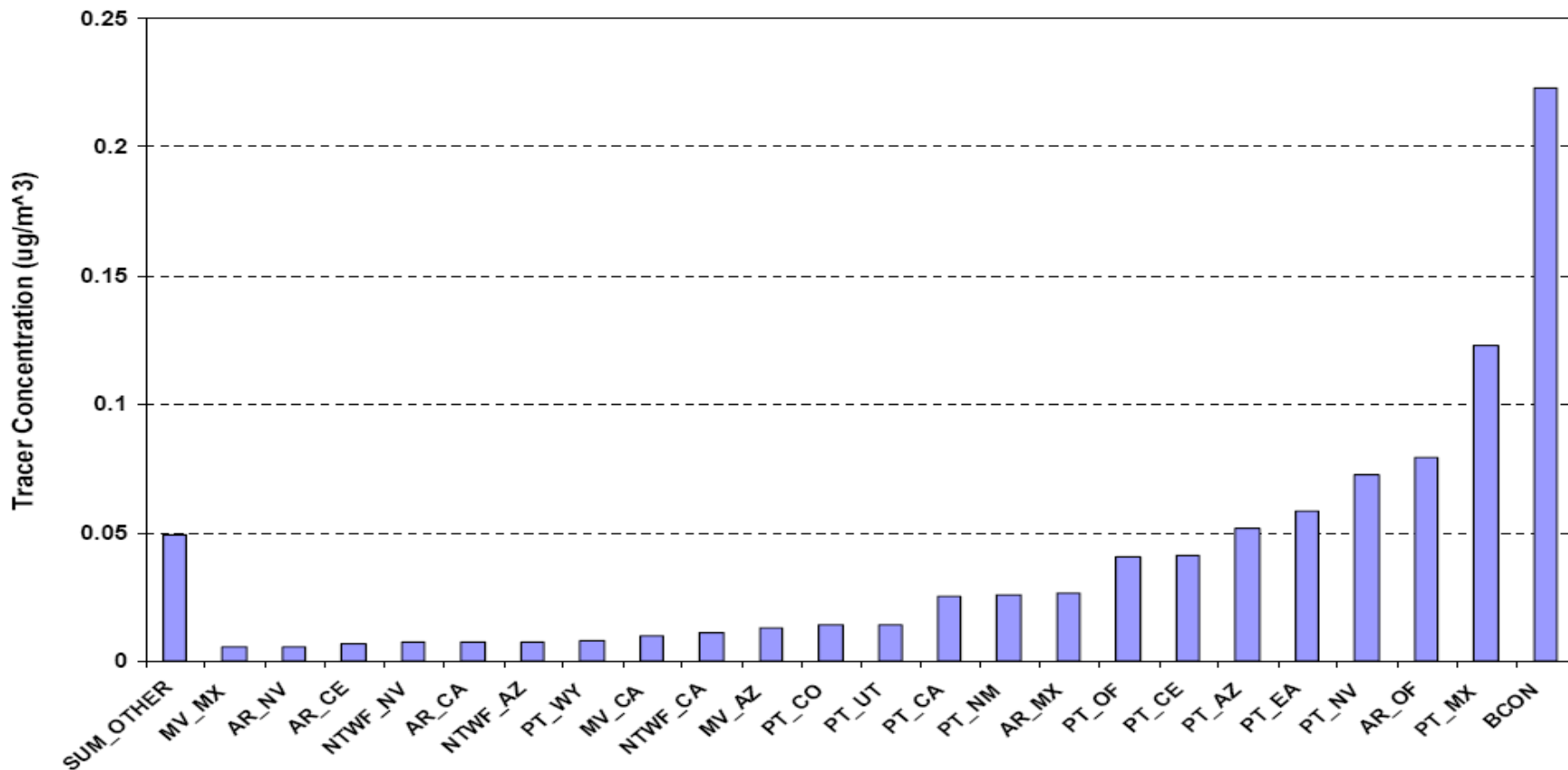


# PSAT Sulfate Contributions at a Receptor Site

## Hance Camp at Grand Canyon NP, AZ (GRCA2)

WRAP Plan02c PSAT Tracers of PSO4: July

Sum of all TRACER Concentration = 0.9257



# Comparison of Rank order of TSSA and PSAT

## July average sulfate contributions at Hance Camp

### CMAQ/TSSA

1. Nevada Point
2. Pacific Offshore Point
3. SOx boundary conc
4. Arizona Point
5. Arizona Mobile
6. Mexico Point
7. Eastern US Point
8. Pacific Offshore Area

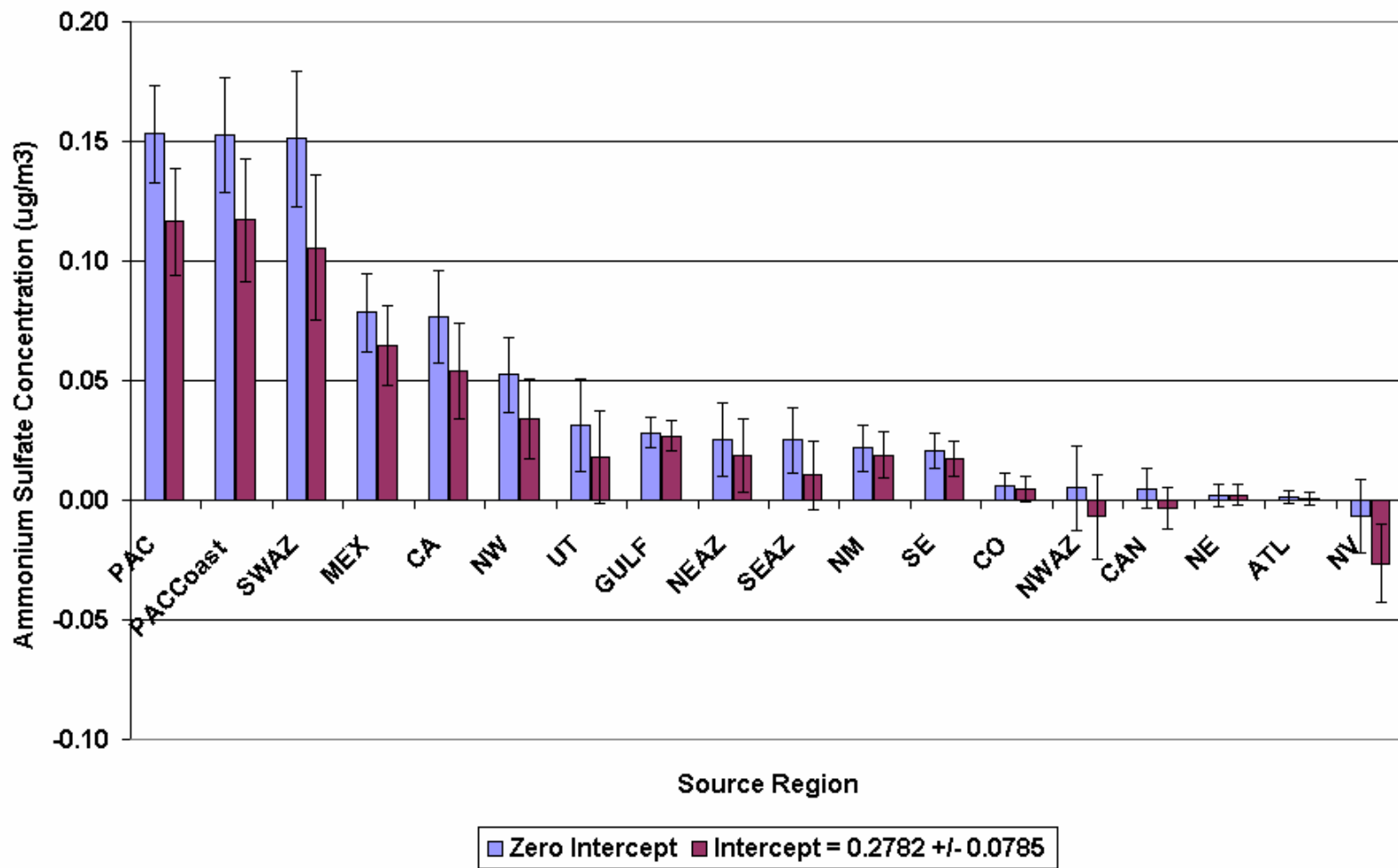
### CAMx/PSAT

1. SOx boundary conc
2. Mexico Point
3. Pacific Offshore Area
4. Nevada Point
5. Eastern US Point
6. Arizona Point
7. Central US Point
8. Pacific Offshore Point

# Sulfate Contributions from HYSPLIT

- DRI HISPLIT modeling results available for annual average. CMAQ/TSSA was run for January and July, so we cannot compare these results.

GRCA2 Sulfate Attribution

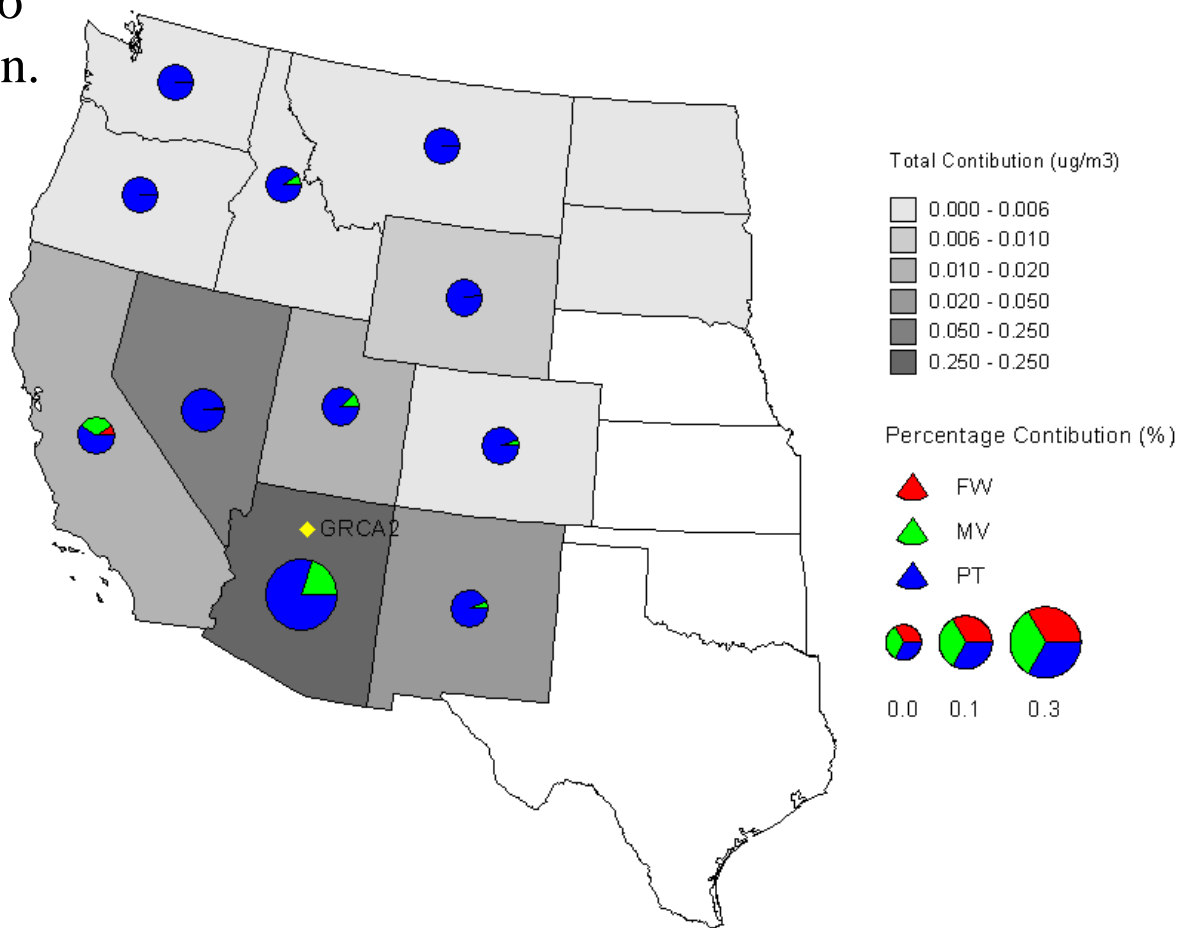


# Sulfate Contributions at a Receptor Site

- Results for January 15, 2002
- Pie charts and color show contribution of each state to sulfate at the Grand Canyon.

TSSA Source Contribution To GRCA2 for Day 1/15/2002

[ East = 0.02      Mex = 0.14      BCONC = 0.06  
[ Can = 0.0029      Not Tagged = 0.05      ICONC = 0



# Conclusions

- Similar results among different methods for source apportionment.
- Most significant difference is the boundary condition contributions from TSSA and PSAT
  - larger BC contribution in PSAT, further analysis of advection and dispersion in CMAQ and CAMx needed to investigate this.

# Summary (1)

- Developed and tested a new source apportionment algorithm in CMAQ using reactive tracers to track the chemical transformation and transport of emissions from selected emissions source categories or regions.
- Can be used to estimate contributions of emissions either from a single source or from selected classes of sources, grouped by source category and/or by region.
- Useful for evaluating the contribution of an small sources that cannot be evaluated in “brute force” sensitivity simulations.

## Summary (2)

- CMAQ/TSSA performed well in comparisons with model brute force sensitivity runs and with back trajectory modeling studies.
- CMAQ TSSA results were also similar to CAMx PSAT, although CAMx PSAT had larger contributions from boundary conditions.
- These results are expected to be useful to air quality managers and scientists both at regulatory agencies and at the regulated industries for evaluating the contribution and importance of individual emissions sources.



# Future Work

- Implement TSSA for organic aerosols and ozone.
- Scalability in supporting other modeling domains
  - Create corresponding source area mapping files
  - county domains.